Sub-Shot-Noise Magnetometry with a Correlated Spin-Relaxation Dominated Alkali-Metal Vapor

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Spin noise sets fundamental limits to the precision of measurements using spin-polarized atomic vapors, such as performed with sensitive atomic magnetometers. Spin squeezing offers the possibility to extend the measurement precision beyond the standard quantum limit of uncorrelated atoms. Contrary to the current understanding, we show that even in the presence of spin relaxation, spin squeezing can lead to a significant reduction of spin noise, and hence an increase in magnetometric sensitivity, for a long measurement time. This is the case when correlated spin relaxation due to binary alkali-atom collisions dominates independently acting decoherence processes.

Quantum noise due to the fundamental quantum-mechanical uncertainties of physical observables sets the standards quantum limits [1] (SQL) for the accuracy of any quantum measurement. Spin-projection noise or spin noise [2], in particular, poses a fundamental limit to the measurement precision using an ensemble of spin systems, be it the actual spin angular momentum of alkali-metal atoms employed, for example, in sensitive magnetometers [3], or other two-level systems such as those involved in atomic clocks [4]. Spin noise of an ensemble of uncorrelated atoms leads to a fundamental noise level that scales as $1/\sqrt{N}$, where N is the number of atoms participating in the measurement process. The creation of quantum correlations between the atoms has emerged as a possibility of extending the measurement precision beyond the SQL of uncorrelated ensembles. Spin squeezing refers to multi-particle quantum states of the system in consideration which exhibit this suppression of quantum noise in spectroscopic measurements [5]. Several theoretical proposals describing ways to create spin squeezing have appeared [6], but so far, spin squeezing has been experimentally demonstrated in systems where decoherence is negligible, i.e. in a cold cesium vapor [7, 8] and in a low density thermal cesium vapor [9]. The motivation for this work is the possibility of enhancing the magnetic sensitivity of atomic mangetometers [10] employing high density alkali-metal vapors by creating spin-squeezed states. These devices have several applications [11, 12] which would benefit from an increased sensitivity beyond the relevant SQL.

However, it was recently conjectured [13] that spin squeezing is of little use in the presence of spin relaxation, leading to sub-SQL magnetic sensitivities only for an impractically short measurement time. This would be detrimental since, unlike laser-cooled atomic ensembles, in high density thermal atomic vapors used in atomic magnetometers spin relaxation is a dominant effect. A similar result was derived for the case of improving frequency standards by use of entanglement [14].

In this Letter we show that spin squeezing does actually lead to a sub-SQL spin noise level and enhanced magnetic sensitivity even in the presence of spin relaxation, and for long measurement times. Using quantum state diffusion theory [15], which naturally reflects the fluctuation-dissipation theorem for the collective atomic spin of uncorrelated atoms, we demonstrate the intimate connection between spin noise and spin relaxation. Hence we find that there is no additional noise due to spin relaxation as suggested in [13], where spin relaxation was treated independently of spin noise. We then show that the dominant relaxation mechanism in a dense alkali-metal vapor, i.e. binary alkali-metal atom collisions, preserves the ensemble quantum correlations, allowing an enhanced measurement precision for a time on the order of the spin relaxation time. We also identify the opposite limit, in which independently acting decoherence mechanisms, if dominant, do actually lead to the conclusions reported in [13, 14].

The physical system we will be considering is a thermal ensemble of alkali-metal atoms confined in a cell. The atoms are initially spin-polarized along the $\hat{\mathbf{x}}$-axis, so that $\langle s_x \rangle = 1/2$, where $\mathbf{s}$ denotes the atom's electron spin. A small magnetic field to be measured is applied along the $\hat{\mathbf{z}}$-axis, and induces a precession of the spins, observed for a measurement time τ. The transverse spin-polarization thus produced can be detected [16] via Faraday rotation of an off resonant probe laser's polarization measured with, e.g., a balanced polarimeter. In a dense alkali-metal vapor the transverse spin relaxation, or spin decoherence, is dominated by two kinds of binary collisions, namely spin-exchange and spin-destruction collisions [17], with respective rates $1/T_{\rm se}$ and $1/T_{\rm sd}$, proportional to the atom density. Both are "sudden" with respect to the nuclear spin and tend to reduce the density matrix [18] $\rho = \phi + \mathbf{a} \cdot \mathbf{s}$ (with ϕ and $\mathbf{a}$ being nuclear operators) to the part ϕ without electronic spin-polarization, i.e. $d\rho/dt = (\phi - \rho)/T_2$, where $1/T_2 = 1/T_{\rm se} + 1/T_{\rm sd}$. At very low magnetic field, which will be assumed hence-

forth, relaxation due to spin-exchange is suppressed [19], and $1/T_2 = 1/T_{sd}$. The time evolution of the density matrix can be written in the Lindblad form

$$\frac{d\rho}{dt} = -i[\mathcal{H}_g, \rho] + \sum_{j=1}^{3}\left(L_j\rho L_j^\dagger - \frac{1}{2}L_j^\dagger L_j\rho - \frac{1}{2}\rho L_j^\dagger L_j\right) \quad (1)$$

where $\mathcal{H}_g$ is the ground state hamiltonian and the three Lindblad operators are

$$L_1 = \frac{1}{\sqrt{2T_2}}s_+, \quad L_2 = \frac{1}{\sqrt{2T_2}}s_-, \quad L_3 = \frac{1}{\sqrt{T_2}}s_z \quad (2)$$

It can be shown [20] that the change of a quantum state $|\psi\rangle$ after the elapse of a time interval dt is given by the quantum state diffusion equation

$$|d\psi\rangle = -idt\mathcal{H}_g|\psi\rangle$$
$$+ \sum_{j=1}^{3}\left(\langle L_j^\dagger\rangle_\psi L_j - \frac{1}{2}\langle L_j^\dagger\rangle_\psi\langle L_j\rangle_\psi - \frac{1}{2}L_j^\dagger L_j\right)|\psi\rangle dt$$
$$+ \sum_{j=1}^{3}(L_j - \langle L_j\rangle_\psi)|\psi\rangle d\eta_j \quad (3)$$

where the first term represents the hamiltonian evolution, the second dissipation, and the third the stochastic fluctuations, described by the statistically independent complex Wiener processes $d\eta_j$, with $j=1,2,3$, i.e. $d\eta_i d\eta_j^* = dt\delta_{ij}$. Since the nuclear spin plays no fundamental role in the following considerations, we will consider an ensemble of N spin-1/2 particles. We furthermore assume the the probing laser is far enough off resonance that we can neglect measurement-induced backaction on the spins [21], that is, we are going to only consider spontaneous spin noise and its effect on measurement precision. This can be done since the probe polarization rotation scales with the probe laser detuning Δ as $1/\Delta$, whereas the measurement strength [21] scales as $1/\Delta^2$. The effects of probe photon shot-noise and scattering have been treated in [13]. From (3) it follows [22] that the expectation value $\langle s_y\rangle$, which is the measured observable, obeys an Ornstein-Uhlenbeck stochastic process,

$$d\langle s_y\rangle = \omega_L\langle s_x\rangle_0 dt - \frac{dt}{2T_2}\langle s_y\rangle + \frac{d\eta}{\sqrt{4T_2}}, \quad (4)$$

where ω_L is the Larmor frequency and now $d\eta$ is a real Wiener process [23], i.e. a normal random variable with zero mean and variance dt. Defining the ensemble transverse spin as $S_y = \sum_{i=1}^{N} s_y^{(i)}$, it follows that for N uncorrelated atoms

$$d\langle S_y\rangle = \omega_L\langle S_x\rangle_0 dt - \frac{dt}{2T_2}\langle S_y\rangle + \Delta S_y\frac{d\eta}{\sqrt{T_2}}, \quad (5)$$

where $\langle S_x\rangle_0 = N\langle s_x\rangle_0$ and $\Delta S_y = \sqrt{N/4}$ is the coherent spin state (CSS) uncertainty. The spectrum of the

transverse spin fluctuations follows a Lorentzian distribution centered at the origin (at zero magnetic field) with a width equal to $1/2T_2$, which also sets the bandwidth of the magnetometric measurement [24]. As is evident from (5), transverse spin dissipation and spin noise are intimately related, both being described by one and the same parameter, T_2. The reason that spin noise sets the SQL for a magnetic field measurement using a collision-dominated alkali-metal vapor is that atomic collisions and the associated relaxation continuously redistribute the variance of the ensemble transverse spin, i.e. even in the infinite time limit when any initial nonzero expectation $\langle S_y\rangle$ has decayed away, $\langle S_y\rangle$ has a non-zero power spectrum extending to $1/2T_2$. This forms the basis of spontaneous spin noise spectroscopy [22, 25]. In contrast, in the case of laser-cooled collision-less atomic vapors [7], the power spectrum of $\langle S_y\rangle$ has only a zero-frequency component, which exhibits a shot-to-shot distribution around zero with an uncertainty $\sqrt{N/4}$ characterizing the CSS [7]. We now assume that the atomic ensemble has been spin-squeezed, i.e. the spin-squeezing parameter [2] $\xi = \sqrt{N}\Delta S_y/\langle S_x\rangle < 1$. That is, as in [26], we assume that a spin squeezing hamiltonian has been applied before probing the precessing spins. In the presence of spin squeezing, however, the spin uncertainties of individual atoms do not simply add in quadrature, as there are negative pairwise correlations [26] that have to be accounted for:

$$C_{yy}^{ij} \equiv \langle s_y^{(i)}s_y^{(j)}\rangle - \langle s_y^{(i)}\rangle\langle s_y^{(j)}\rangle = \frac{\xi^2 - 1}{4N}, \quad (6)$$

leading again to Eq. (5), but now

$$\Delta S_y = \xi\sqrt{N/4} \quad (7)$$

This is the first main result of this work. Spin relaxation obviously leads to dissipation of a non-zero expectation value $\langle S_y\rangle$. At the same time it is manifested through spin noise, i.e. the fluctuations of $\langle S_y\rangle$ around its mean value, described by the third, stochastic term of Eq. (5). There is no other noise source due to spin relaxation. We emphasize that these fluctuations are driven by atomic collisions and not by photon noise coupling into $\langle S_y\rangle$, as is the case with strong measurements [27] of the collective spin of laser-cooled vapors. The magnitude of these fluctuations, given by Eq. (7) is indeed reduced if $\xi < 1$ and a sub-SQL sensitivity can be achieved for a measurement time $\tau \approx T_2$. Indeed, from (5) it follows that the maximum value of $\langle S_y\rangle$ will be $\langle S_y\rangle_\tau = \omega_L 2T_2\langle S_x\rangle_0$ at $\tau \approx T_2$. The noise acquired at time $\tau = T_2$ due to the stochastic term of the evolution equation (5) will on the average be $\delta\langle S_y\rangle = \xi\sqrt{\frac{N}{4T_2}}\sqrt{T_2} = \xi\sqrt{N}/2$. Thus the sensitivity limit for measuring a small frequency ω_L will be (neglecting factors of 2)

$$\delta\omega_L = \frac{\delta\langle S_y\rangle}{\partial\delta\langle S_y\rangle/\partial\omega_L} = \frac{\xi}{T_2\sqrt{N}} \quad (8)$$

By averaging n such measurements for a total measurement time $T = nT_2$, we get

$$\delta\omega_L = \frac{\xi}{\sqrt{NT_2 T}} \qquad (9)$$

For the case of uncorrelated atoms, $\xi = 1$, we recover the well-known shot-noise limit. In the above derivation we have neglected the decay and fluctuations of $\langle S_x \rangle$. This is allowed, since the decay of $\langle S_x \rangle$ during the measurement time T_2 will change our estimate by a factor of order 1, and the fluctuations of $\langle S_x \rangle$, of order $\sqrt{N}$, leak into $\langle S_y \rangle$ but are diminished by the spin rotation angle, $\omega_L T_2$. For a magnetic field magnitude in the fT range and a spin coherence time $T_2 \approx 1$ ms, this factor is of order 10^{-8}, and for all practical purposes, is negligible compared to the actual spin noise of $\langle S_y \rangle$, of magnitude $\xi\sqrt{N}$.

In the following we are going to justify the previous assertions in more detail. The reason that spin noise is reduced even in the presence of relaxation is that the decoherence mechanism we are considering is not acting on every atom independently, resulting in a change $d(\Delta S_y)^2 = 0$ to first order in dt. Indeed, in a binary collision between alkali-metal atoms the dominant interaction leading to relaxation is the so-called spin-axis interaction [17, 28], described by a hamiltonian of the form $\mathcal{H}_{\text{sa}} = \lambda(3\sigma_\zeta \sigma_\zeta - 1)$, where σ_ζ is the projection of the total electron spin of the colliding atoms on the internuclear axis and λ is a coupling constant. This represents correlated decoherence [29], the effect of which is that it preserves spin correlations. To prove that, we consider two spin-1/2 particles in the triplet sub-space [26] state $|\psi\rangle = \alpha|00\rangle + \beta(|01\rangle + |10\rangle)/\sqrt{2} + \gamma|11\rangle$ colliding along some axis $\hat{\boldsymbol{\eta}}$. The spin-axis hamiltonian reduces to $\mathcal{H}_{\text{sa}} \sim \lambda s_\eta^{(1)} s_\eta^{(2)}$. The change in the initial density matrix $\rho = |\psi\rangle\langle\psi|$ induced by $\mathcal{H}_{\text{sa}}$ will be $d\rho = -i[\mathcal{H}_{\text{sa}}, \rho]dt$. We then calculate the correlation C_{yy}^{12} (or concurrence [30]) in the state ρ, and find $C_{yy}^{12} = (\beta^2 - 2\alpha\gamma)/4$. Whereas the change in the expectation value $\langle S_y \rangle = \langle s_y^{(1)} + s_y^{(2)} \rangle$ is found to be proportional to λdt, the change in the correlation dC_{yy}^{12} is found to be proportional to $(\lambda dt)^2$. This means that $\mathcal{H}_{\text{sa}}$ induces dissipation, as expected, but preserves two-body quantum correlations of the spins.

We now turn to the many-particle spin-squeezed state introduced in [26], $|\Psi_{ss}\rangle = \sum_{l=0}^{N} c_l |\phi_l^{\text{perm}}\rangle$, where $|\phi_l^{\text{perm}}\rangle = \sum_{\text{perm}} |1^{\otimes l} 0^{\otimes N-l}\rangle$ is a permutation symmetric state with l 1's and $N - l$ 0's, $c_l = i^l b_l$ and b_l are given in [26]. We take dt to be the duration during which particles 1 and 2 have interacted through $\mathcal{H}_{\text{sa}}$, resulting in a state change $|d\Psi_{ss}\rangle$. The change in the ensemble variance is $d(\Delta S_y)^2 = \langle d\Psi_{ss}|S_y^2|\Psi_{ss}\rangle + \langle \Psi_{ss}|S_y^2|d\Psi_{ss}\rangle$. Since $\langle S_y \rangle \approx 0$, it follows that $S_y|\Psi_{ss}\rangle$ is almost perpendicular to $|\Psi_{ss}\rangle$ and therefore $S_y^2|\Psi_{ss}\rangle$ is proportional to $|\Psi_{ss}\rangle$. Thus $d(\Delta S_y)^2 \sim \mathfrak{Re}\{\langle d\Psi_{ss}|\Psi_{ss}\rangle\}$. By use of their symmetry, it is easily seen that the change induced by $\mathcal{H}_{\text{sa}}$

in the states $|\phi_l^{\text{perm}}\rangle$ is

$$|\delta\phi_l\rangle = -i\lambda dt \left(\alpha|\chi_{l-2}\rangle + \beta|\chi_l\rangle + \alpha|\chi_{l+2}\rangle \right) \qquad (10)$$

where the real coefficients α and β depend on the particular collision trajectory and the states $|\chi_m\rangle$ contain a subset of the terms of the corresponding states $|\phi_m^{\text{perm}}\rangle$, with $m = l, l\pm 2$, and hence $\langle \chi_m|\phi_m^{\text{perm}}\rangle$ is a real number. Thus the overlap $\langle d\Psi_{ss}|\Psi_{ss}\rangle \sim i\lambda dt(\alpha c_{l-2} + \beta c_l + \alpha c_{l+2})c_l^*$, and therefore, since $b_l \approx b_{l\pm 2}$ for large l, $\mathfrak{Re}\{\langle d\Psi_{ss}|\Psi_{ss}\rangle\} = 0$. We have thus proved in the most general way that correlated relaxation preserves the ensemble variance $(\Delta S_y)^2$, i.e. $d(\Delta S_y)^2 = 0$ to first order in dt.

On the contrary, independently acting decoherence processes will tend to reduce the ensemble correlations during the measurement. Based on the above considerations, when an atom decoheres independently, the change in $|\phi_l^{\text{perm}}\rangle$ is proportional to a linear combination of $|\chi_{l\pm 1}^{\text{perm}}\rangle$, and that leads to a non-zero real part of the overlap $\langle d\Psi_{ss}|\Psi_{ss}\rangle$, which is of order dt. The consequence of the decay of $(\Delta S_y)^2$ can be simply described with an effective squeezing parameter $\xi' > \xi$. Indeed, if we assume that there are two decoherence mechanisms, one acting independently on each atom and one preserving $(\Delta S_y)^2$, with respective rates $1/T_{\text{nc}}$ and $1/T_{\text{c}}$ (obviously $1/T_2 = 1/T_{\text{nc}} + 1/T_{\text{c}}$), using Eqs. (4) and (6) we again arrive at (7), but with ξ replaced by

$$\xi' = \sqrt{\frac{\xi^2 + T_{\text{c}}/T_{\text{nc}}}{1 + T_{\text{c}}/T_{\text{nc}}}} \qquad (11)$$

If $T_{\text{c}}/T_{\text{nc}} \gg 1$, we get $\xi' \approx 1$ and recover the case of uncorrelated spin noise described by (5), with $T_2 = T_{\text{nc}}$. This limit corresponds to the results obtained in [13, 14, 26] where entanglement in the presence of decoherence is shown not to offer any increase in measurement precision beyond the uncorrelated ensemble case. On the other hand, since there will always be an independently acting relaxation mechanism, such as atom collisions with buffer gas atoms or container walls, in the limit that $T_{\text{c}}/T_{\text{nc}} \ll 1$ there is no point in attempting to reduce ξ^2 below this ratio. For example, using the spin-destruction cross sections [3] for K-K and K-He collisions, we find that for a potassium density of $[\text{K}]=10^{15}$ cm^{-3} in the presence of 1 atm of helium buffer gas, it is $T_{\text{c}} \approx 1$ ms and $T_{\text{nc}}/T_{\text{c}} \approx 50$, suggesting, were it experimentally feasible, the need for a squeezing parameter no smaller than $\xi \approx 0.1$. The above considerations imply a new fundamental sensitivity limit for a frequency measurement. Based on (9) and assuming $\xi \approx \sqrt{T_{\text{c}}/T_{\text{nc}}}$ we find

$$\delta\omega_L = \frac{1}{\sqrt{NT_{\text{nc}}T}}, \qquad (12)$$

but now $T_2 = T_{\text{c}} \ll T_{\text{nc}}$. This is the second main result of this work. Essentially, with spin squeezing we manage to suppress the effect of correlated spin relaxation on

the measurement precision. Undoubtedly, the measurement precision still scales as $1/\sqrt{N}$. However, N can now be made as large as is practically possible, since T_{nc} is atom-density independent. Therefore, $\delta\omega_L$ can be made arbitrarily small, in contrast to the uncorrelated-atoms case, where $\delta\omega_L$ saturates at high densities since the relaxation rate $1/T_c$ is proportional to the atom density. Furthermore $1/T_{nc}$ represents relaxation due to "technical" imperfections, such as relaxing cell walls, that is not in any fundamental way prevented from reaching small values.

Since is is straightforward to show that independently acting decoherence tends to reduce $(\Delta S_y)^2$ to the uncorrelated-atoms value of $N/4$ at a rate $1/T_{nc}$, we can more formally arrive at Eq. (12) by solving the set of equations

$$d\langle S_y\rangle = \omega_L\langle S_x\rangle_0 e^{-t/T_2}dt - \frac{dt}{2T_2}\langle S_y\rangle + \Delta S_y\frac{d\eta}{\sqrt{T_2}} \quad (13)$$

$$d(\Delta S_y)^2 = -\left((\Delta S_y)^2 - \frac{N}{4}\right)\frac{dt}{T_{nc}} \quad (14)$$

where the decay of $\langle S_x\rangle$ has also been included for completeness and initially $\Delta S_y(t=0) = \xi\sqrt{N/4}$. For a measurement time T_c, the above equations lead to a sensitivity $\delta\omega_L = 1/\sqrt{NT_cT_{nc}}$. Averaging $n = T/T_c$ such measurements leads again to (12).

In conclusion, we have shown that by suppressing spin noise due to correlated spin relaxation, spin squeezing results in an increased measurement precision during a long measurement time in a collision-dominated alkali-metal vapor. Similar comments apply to the case of atomic clocks employing thermal alkali-metal vapors. The spin-exchange interaction during binary collisions, which is the main decoherence mechanism in these clocks, and of the form $\mathcal{H}_{se} \sim \mathbf{s}_1\cdot\mathbf{s}_2$, shares the same property with the spin-axis interaction, i.e. it results in correlated decoherence. Spin squeezing the clock transition could thus significantly boost the clock performance of frequency standards employing thermal alkali-metal vapors.

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